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Appl. No. 10/538,136

Reply to Office Action of February 12, 2009

REMARKS/ARGUMENTS

In the last Action the Examiner took the position of disagreeing with applicants' arguments. Therefore the rejection was continued. There is attached hereto a translation of Murata to help applicants better explain differences between the invention and the art.

The present invention as required in the claims is directed to a multilayer ceramic electronic part having an external electrode(s) which is characterized in that the external electrode is formed by curing a specific thermosetting conductive paste. The specific thermosetting conductive paste comprises conductive particles having a high melting point of 400 °C or more (hereinafter "high melting point conductive particle"), metal powder having a melting point of 300 °C or less (hereinafter "low melting point metal powder") and thermosetting resins(s). The claims require that the low melting point metal powder is present in an amount by weight based on the total weight of the high melting point conductive particle and the low melting point metal powder, of from 5% to 17.6%. By forming an external electrode(s) using such paste, bonding property between an internal electrode(s) and the

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external electrode is improved and excellent electric property and high reliabilities can be obtained. Support can be found in Table 4 of the present specification. Example 1f (low melting point metal powder is present in 5 wt%) and Example 1 (17.6 wt%) have electric capacitances greater than those of Example 1e (1 wt%) and Example 1g (25 wt%). Examples 1f and 1 have  $\tan \delta$  smaller than those of Examples 1e and 1g. Examples 1f and 1 also have high bonding strength.

Shioya is cited to show a multilayer ceramic electronic part having an external electrode made from thermosetting paste. The invention disclosed in Shioya is directed to a terminal electrode for electronic parts. The terminal electrode comprises metallic particles and resin (Claim 1). The metallic particles have different composition between internal frame portion (internal portion) and portion near the surface of the metallic particle. The internal frame portion of the metallic particle is a substance with a melting point of 600°C or more and the portion near the surface of the metallic particle is a substance with a melting point of 500°C or less (Claim 1). Shioya uses a conductive paste comprising metallic particle with a melting point of 600°C or more which is coated by a substance with a melting point of 500°C or less in order to soften only the portion near the surface of the metallic particle when heat is

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applied ([0011]). For example, silver powder, copper powder and nickel powder each having indium or tin coating are used in Examples of Shioya [0013]. In view of this, Shioya teaches away from using a mixture of high melting point conductive particles and low melting point metal powder. The low melting point metal is on the surface of the high melting point powder e.g. by electroplating. It is a matter of course that there is no description about a mixture of high melting point conductive particle and low melting point metal powder in Shioya.

Further, Shioya is silent about by what amount of the substance with a melting point of 500°C or less (portion near the surface) should the substance with a melting point of 600°C or more (internal portion) be coated. Nor is there any suggestion of an appropriate amount.

It is submitted that a coated particle and a mixture of particles are different in behavior when it is exposed to heat. There is no suggestion that they are interchangeable and, in view of this difference, it would not be obvious to interchange them. Even if Shioya describes an amount of the substance with a melting point of 300°C or less it would not show or suggest a mixture of particles.

As stated above, Shioya describes nothing about using a mixture of high melting point conductive particle and low

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melting point metal powder, and is silent about using the low melting point powder in an amount of 5 wt% to 17.6 wt%. Thus, the present invention differs from Shioya. The present invention is not obvious over Shioya. Relying on Murata to show the particles does not render the present invention obvious.

Combining Murata with Shioya appears to be improper. This was explained in the earlier response. Shioya belongs completely different technical field from Murata as explained below.

First of all, Murata only discloses a conductive paste used for forming hot-wire heater on the rear window of a car defogger. Murata is silent about a conductive paste used for print circuit board. [The description of Derwent-abstract has no ground. Please refer to the full translation of Murata.] Meanwhile, Shioya discloses a conductive paste used for a terminal electrode for electronic parts. Shioya belongs to a completely different technical field from Murata.

In the final OA, the Examiner relies on the teaching in Murata for the purpose of presenting the limitation of "the metal powder having a melting point of 300°0 or less is present in an amount by weight based on the total weight of

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said conductive particles having a high melting point and  
said metal powder having a melting point of 300°C or less,  
of from of 5 % to 17.6%".

However, one attempting to solve the objects of the  
present invention, would not look to Murata. Murata, as  
noted above, is for a much different purpose. The type and  
amount of metal particle in a paste is determined  
considering the property required for the product obtained  
by using the paste, compatibility between the metal  
particle and the other components of the paste, method of  
applying the paste (heating temperature, etc.). It is not  
reasonable to take out of context only about metal powder  
apart from the other disclosure.

As examples of differences in properties, it is noted  
that a conductive paste of Murata is used to prepare a  
conductive film which is a hot-wire heater on the rear  
window of a car defogger. Resistant value of the conductive  
film is important in view of function as a hot-wire heater,  
and type and amount of metal particle is selected so as to  
stably obtain desired resistant value in Murata (page 3,  
line 5 of the same). A resistance to abrasion as well as a  
resistance to weather is also important, and type and  
amount of metal particle is selected so as to improve these

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properties in Murata (page 3, lines 6 to 8 of the same).

Meanwhile, a terminal electrode is prepared by a paste in Shioya, and thus electric capacitance, dielectric loss and bonding property with internal electrode are looked over in Shioya. Criteria for selecting metal powder are remarkably different between Murata and Shioya. Particularly, in Murata, Sn powder is categorized as weather-resistance reinforcement agents (page 5, lines 4 to 6 of the same), and there is no suggestion that Sn powder contribute to improve properties of a terminal electrode.

Further, the paste disclosed in Murata is a sintering type which contains glass frit as a binder component, and is heated to about 700°C after application (Claims, Example). Meanwhile, the paste in Shioya contains thermosetting resin, and is heated to about 200 to 250°C after application (Example). The paste in Murata is applied to glass substrate. On the other hand, the paste in Shioya is applied to ceramic body. Components of the pastes, subject of the application and method of application are remarkably different between Murata and Shioya.

As stated above, Murata belongs to a technical field which is different from the which Shioya belongs. The properties required are much different. Normally one does not look to a resistance material for electric circuit connectors ([0007]) of

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Murata refers to "excellent in" conductivity). Thus it is unreasonable to combine Murata with Shioya.

Further, as stated above, the particle disclosed in Shioya is a coated particle (i.e., metallic particle with a melting point of 600°C or more which is coated by a substance with a melting point of 500°C or less), but not a mixture (i.e., mixture of metal particle with high melting point and metal particle with low melting point). Shioya teaches away from such mixture by requiring a coated particle. Thus, the combination does not render the present invention obvious.

In the final OA, it is pointed out that the amount of Sn powder of Sample 3 in Table 2 of Murata is 8.92 wt% (page 4, line 11 to page 5, line 18 from the bottom). However, the amount of Sn powder of Samples 9 and 12 in the same table is only 0.06 wt%. It is unreasonable to select only Sample 3, while ignoring Samples 9 and 12. Also, Murata needs the internal portion to be covered with the low melting metal to reduce deterioration by requiring that less expensive (and easily oxidized) material be protected by a coating [003], [0070], [0009]. Thus substituting the powder mixture prevents the object of Murata since there would be no protective coating.

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The present invention is also rejected as obvious over Seo in view of Fukuda (page 8, line 4 from the bottom to page 12, last line).

Seo is cited to show thermosetting conductive past with high and low melting point particles. Although the invention disclosed in Seo is directed to an electroconductive adhesive, Seo does not disclose a thermosetting electroconductive adhesive, or its use for forming an external electrode of electronic parts.

It is noted that the electroconductive adhesive in Seo is characterized by its reworkability (paragraph 3). In Seo, "reworkability" means properties such that if circuit has insufficient conductivity once after attached by an conductive adhesive, the component which forms such portion can be removed by known method, and reattached to correct defects (paragraph 3). Further, "rework" means that a component attached to a member is removed by heating the member to which the component is attached at a temperature above glass transition temperature of thermoplastic resin and below a temperature which would deteriorate the member be heated, or by pulling the component with slight force after treatment by solvent to lower shear strength to 3kgf or less (paragraph 23). Since the



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electroconductive adhesive of Seo is used for connection of complicated circuit board, etc., reworkability is important.

In contrast, meanwhile, an external electrode of electronic parts (as claimed herein) is required to have excellent bonding property with an internal electrode. Thus, it is not practical if an external electrode is easily removed when heat is applied. Further, properties concerning mounting to substrate or plating are also important for an external electrode. Usually, an external electrode is mounted to substrate by soldering. Accordingly, heat of molten solder affects the external electrode. It is impossible to obtain an end-product if the external electrode is easily removed thereby. It is therefore submitted that one of ordinary skill in the art would not consider the Seo disclosure when making the present invention. The properties of Seo are contradicted for the present invention.

In Examples of the present specification, an external electrode of multilayer ceramic electronic element was plated and then solder jointed with the substrate, and the bonding strength of thus obtained test sample was determined. In the present invention, the bonding strength was high and hardly decreased after heat-cycles, and excellent reliability is

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maintained. These properties are opposite the properties which are the object of Seo.

As stated above, the electroconductive adhesive of Seo has properties which are not acceptable to an external electrode of multilayer ceramic electronic element. Thus, Seo is not an appropriate prior art reference.

Further, the electroconductive adhesive of Seo comprises thermoplastic resin to ensure reworkability (paragraphs 16 and 17). In general, thermosetting conductive paste including the present invention never softens by reheat, once it is set by heat. Thus, it is not appropriate for ensuring reworkability. Accordingly, the electroconductive adhesive of Seo cannot be regarded as thermosetting conductive paste. Moreover, Seo teaches away from thermosetting conductive paste.

Combining Fukuda does not bridge the missing teaching. Also, for the reasons detailed above that Seo is an inappropriate reference, it is not obvious to combine Fukuda with Seo. The invention disclosed in Fukuda is directed to a chip-like electronic part. As stated above, the electroconductive adhesive of Seo has properties which are not acceptable to an external electrode of multilayer ceramic electronic element. Thus, a person skilled in the

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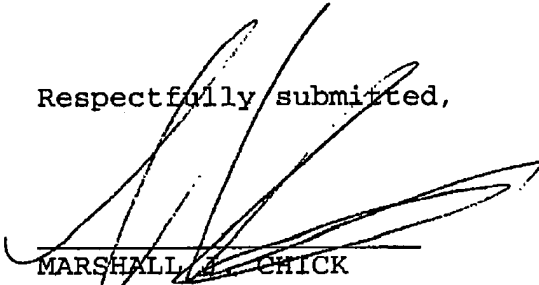
art would never combine Seo with Fukuda, nor look to Seo to provide teaching.

Further, Fukuda is silent about a composition of an electroconductive adhesive used for an external electrode of a chip-like electronic part. There is no motivation to employ the electroconductive adhesive of Seo to prepare the external electrode of a chip-like electronic part of Fukuda. Even when the electroconductive adhesive of Seo is combined, this electroconductive adhesive would have a different composition compared with that of the thermosetting conductive paste of the present invention. Therefore, the present invention is not obvious from a combination of Seo in view of Fukuda nor is the combination appropriate.

In view of the above, the rejections are avoided. Allowance of the application is therefore respectfully requested.

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Enc. Translation of Murata

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TRANSLATION OF  
Document cited  
By the Examiner

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## SPECIFICATION

## 1. Title of the invention

Conductive paste

5

## 2. Claims

A conductive paste comprising:

40 to 87wt% of silver powder;

1 to 10wt% of glass frit;

10

0.5 to 20wt% of at least one powder of Al, Mn, Cu, Zn, Pd, Cd, Sn, Sb, Pt, PB, and

Bi; and

10 to 40wt% of an organic varnish, wherein:

(a) at least one of Al, Mn, Ni, Pd, Pb, and Bi is 0.5 to 13wt%;

(b) at least one of Cu and Pt is 0.5 to 6wt%; and

15

(c) at least one of Zn, Cd, Sn, and Sb is 0.5 to 10wt%.

## 3. Detailed description of the invention

The present invention relates to a conductive paste for obtaining a conductive film that offers great resistance to abrasion and weather as well as an excellent solderability, and is highly adhesive.

20

An application of the conductive paste is, for example, a wire heater for defogging the rear windshield of a car. Conventionally, such wire heater has been obtained by applying the conductive paste on a glass substrate in a certain pattern, baking it to form a conductive film, electrolytic plating it with copper, and further electrolytic plating with nickel thereon.

25

In this case, a method has been taken wherein: the resistance value is previously adjusted by processing the conductive paste by, for example, dilution to reduce the content rate of silver contained therein, or changing a printing screen mesh; the conductive paste is then applied on the glass substrate by means of the screen printing; the substrate is afterward baked at approximately 700°C, the temperature of reinforcing glasses, to form the conductive film; and the fine adjustment of the resistance value is made at the time of electrolytic plating the substrate with copper and nickel.

30

Carrying out the electrolytic plating process, however, leads to the increase in cost because the manufacturing steps increases, a control device is required for fine adjusting the resistance value, and it is necessary to install and manage the antipollution facility for treating plating waste. Therefore, it has been desired to realize a conductive film without

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having to plating, i.e., a non-plating conductive film.

The present invention is directed to obtain a conductive film capable of meeting the requirement as described above, and to provide a conductive paste characterized by the following features (1) to (6), wherein:

- 5 (1) a resistant value is obtained as desired without plating;
- (2) a resistance to abrasion is offered which is equal or superior to that of the conductive film having the nickel plating on the surface thereof;
- (3) a resistance to weather is offered which is equal or superior to that of the conductive film having the nickel plating on the surface thereof;
- 10 (4) a terminal can be soldered easily;
- (5) high adhesibility after soldering the terminal is provided; and
- (6) a distortion given to the substrate after soldering the terminal is minimized.

15 In order to obtain the above-mentioned features, it is the primary object of the present invention to provide a conductive paste, characterized in that:

- a conductive paste comprising:
  - 40 to 87wt% of silver powder;
  - 1 to 10wt% of glass frit;
  - 0.5 to 20wt% of at least one powder of Al, Mn, Cu, Zn, Pd, Cd, Sn, Sb, Pt, PB, and
  - 20 Bi; and
  - 10 to 40wt% of an organic varnish, wherein:
    - (a) at least one of Al, Mn, Ni, Pd, Pb, and Bi is 0.5 to 13wt%;
    - (b) at least one of Cu and Pt is 0.5 to 6wt%; and
    - (c) at least one of Zn, Cd, Sn, and Sb is 0.5 to 10wt%.

25 The conductive film having, for example, 0.3mm in width and 300mm in length obtained by this conductive paste provides the resistance value ranging from 1.5 to 3.0  $\Omega$ .

30 In this conductive paste, silver, in a powder form having the average particle diameter of 10 $\mu$  or less, is used to obtain the resistance value and baking condition as desired. When the baking temperature is relatively low, the powder having the average particle diameter of 2 $\mu$  or less accelerates the sintering. Metallic silver as well as silver oxide powder offers a good conductive film. In order to obtain the resistance value as desired and maintain the solderability, the paste must contain silver within the range from 40 to 87wt%.

35 A glass frit enhances the bonding strength between the conductive film and the substrate for better solderability, including borosilicate glass such as borosilicate bismuth

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and borosilicate lead, zinc borosilicate glass, borofluoride lead glass, whose average particle diameter thereof is preferably  $5\mu$  or less, and whose softening temperature is preferably 400 to 850°C.

Table 1 shows examples of the composition of the aforementioned glass frit.

5

Table 1

Glass	Composition (wt%)									
frit	PbO	Bi <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	B <sub>2</sub> O <sub>3</sub>	CdO	ZnO	Al <sub>2</sub> O <sub>3</sub>	Li <sub>2</sub> O	CaO	PbF <sub>2</sub>
A	75.0		7.0	15.0			1.0	2.0		
B		85.0	8.0	5.0	3.0			1.0		
C	43.0	43.0	6.0	8.0						
D			10.0	40.0		45.0		1.0	4.0	
E	39.0		10.0	30.0	5.0					16.0

D in Table 1 indicates the glass frit suitable for baking in the reducing atmosphere.

10 The glass frit is limited to 1 to 10wt% because the minimum bonding strength for practical use ( $10\text{kg}/\text{cm}^2$ ) cannot be obtained at less than 1wt%, while good solderability cannot be obtained at above 10wt%.

Among the powders of Al, Mn, Ni, Cu, Zn, Pd, Cd, Sn, Sb, Pt, Pb, and Bi, Al, Mn, Ni, Pd, Pb, and Bi are for obtaining a desired resistance value steadily, and act as resistivity adjustment agents. Al, Mn, Ni, Pd, Pb, and Bi are hereinafter referred to as resistivity adjustment agents. At least one among those resistivity adjustment agents is set to 0.5 to 13wt% because the addition of less than 0.5wt% produces no effect, while a desired resistance value, specifically the resistance value within  $30\Omega$ , cannot be obtained and silver becomes significantly brittle above 13wt%. In the resistivity adjustment agents, Ni, Pb, and Bi are suitable for baking in an oxidative atmosphere, Al and Pd are suitable for baking in a neutral (nitrogen) atmosphere, and Mn is suitable for baking in a reducing atmosphere. If it is desired to bake Al, Pd, and Mn in an oxidative atmosphere, it can be achieved by coating the surfaces of the metal powders with the metal that is not subject to oxidation, such as silver.

25 Cu and Pt are for obtaining the resistance to abrasion that has been achieved by nickel electrolytic plating conventionally, and act as abrasion-resistance reinforcement agents. Cu and Pt are hereinafter referred to as abrasion-resistance reinforcement agents. At least one among those abrasion-resistance reinforcement agents is set to 0.5 to 10wt% because the addition of less than 0.5wt% produces no effect, while silver becomes brittle at above 10wt% and thus the conductive film delaminates inside of the layer. In addition,

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although Cu is required to be baked in the nitrogen atmosphere, coating the surface with the metal that is not subject to oxidation makes it possible to perform the baking in the oxidative atmosphere, as described in the resistivity adjustment agents section.

5 Zn, Cd, Sn, and Sb do not require the plating after baking. They are most important in the conductive pastes, and act as weather-resistance reinforcement agents. Zn, Cd, Sn, and Sb are hereinafter referred to as weather-resistance reinforcement agents.

10 The weather-resistance reinforcement agent prevents abnormal heat generated by the increase of the resistance value due to discoloration of silver. Using the weather-resistance reinforcement agent eliminates the nickel electrolytic plating process that has prevented such phenomenon conventionally. At least one among those weather-resistance reinforcement agents is set to 0.5 to 11wt% because no conductive film with a resistance to weather can be obtained at less than 0.5wt%, while silver becomes brittle significantly at above 10 wt%.

15 Total amount of the resistivity adjustment agent, the abrasion-resistance reinforcement agent, and the weather-resistance reinforcement agent is set to 0.5 to 20wt% because the characteristics that those components have cannot be obtained at less than 0.5wt%, while the conductive film delaminates inside of the layer at above 20wt% and thus good solderability cannot be maintained.

20 While the description has been made wherein the surfaces of the metal powers, Al, Pd, Mn, and Cu, are covered with, as an example, silver, because they are baked in the oxidative atmosphere, the same effect is revealed when the others, Ni, Zn, Cd, Sn, Sb, Pt, Pb, and Bi, are subjected to the process to their surface with silver. Specifically, this leads to a good fitness for silver and high baking density of the silver so as to strengthen the adhesibility.

25 The solid matters described above are dispersively suspended in an organic varnish to be processed to a paste, which is then applied to a substrate by means of painting, printing and the like, followed by being baked to be a conductive film. When the solid matters are mixed with the organic varnish to make a paste, the content of the organic varnish can be changed depending on the application means and the type of the organic varnish, which is preferably within the range of 10 to 40wt%. This is because, at less than 30 10wt%, the content of the solid matter is too high to make a paste, while, at above 40wt%, the film thickness of the conductive film becomes 5 $\mu$  or less, which leads to the poor resistance to abrasion to result in the short life.

35 An example of the organic varnish includes a ethyl cellulose resin dissolved in terpineol or cellsolve. This is suitable for the screen printing.

In addition to the above, the addition of 0.5 to 5.0wt% of organic noble metal



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colloids such as organoplatinum, organorhodium, organogold, and organosilver can promote the baking. The addition out of this range, i.e., less than 0.5wt% produces no effect, while the addition above 5.0wt% makes the baked conductive film brittle, thereby causing the delamination inside of the layer.

5 The present invention will be described below according to Examples.

#### Examples

10 Raw materials, i.e., silver, a glass frit, a resistivity adjustment agent, an abrasion-resistance reinforcement agent, a weather-resistance reinforcement agent, and an organic varnish, were mixed at the composition ratio shown in Table 2 to make a paste. The paste was printed or painted on a glass substrate, baked for three minutes at 700°C, and then cooled rapidly.

The glass frit used was the one having the composition shown in Table 1.

15 Table 2

Sample Number	Composition (wt%)					
	Ag powder	Glass frit	Resistivity adjustment agents	Abrasion-resistance reinforcement agents	Weather-resistance reinforcement agents	Organic varnish
1 *	37	B=8	Al=5	-	Zn=11	39
2 *	42	D=0.5	Mn=0.2 Ni=0.2	Cu=7	-	50.1
3	45	C=7	Pd=6	-	Sn=5	37
4	59	B=5	-	Pt=6	Cd=10	20
5 *	60	E=5	Al=3 Ni=3 Bi=9	Cu=5	Sn=5	10
6	66	D=2	Bi=9	Cu=5 Pt=1.5	Sb=5	23
7	72	C=2	Ni=1	-	Zn=2 Sb=3	20
8 *	76.5	A=12	-	Pt=0.2	Sb=0.2	11.1
9	78.5	E=1	-	-	Sn=0.5	20
10 *	79	C=1	-	-	-	20
11	80.5	B=4	Pb=4	-	Cd=0.5	11
12	86.5	A=1	-	-	Sn=0.5	12
13 *	90	A=1.5	Ni=1	-	-	7.5

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The conductive film obtained above was measured regarding the resistance value, solderability, adhesibility, and the resistance to abrasion and weather thereof. The results are shown in Table 3.

5 Each characteristic was measured under the conditions as follows:

Resistance value: measured in the pattern having 0.3mm (width) x 300mm (length);

Solderability:

10 "excellent" – Solder was attached perfectly to the conductive film. Solder wettability is excellent.

"good" – 80% of solder was attached to the conductive film.

"acceptable" – Solder was attached perfectly when the conductive film was polished with steel wool.

"bad" – Solder was not attached at all.

15 Adhesibility: the value obtained by soldering an L-shaped soldering annealed copper wire to the conductive film whose area is 2mm x 2mm, followed by conducting the tensile test thereto. The adhesibility of above 1kg/4mm<sup>2</sup> is required for practical use.

20 Resistance to abrasion: A line of the conductive film of 0.8mm in width was baked on the glass substrate to subject to the abrasion test by means of the abrasion testing machine of Taber, Abraser Model 503. The determination was done by the number of rotation when the conduction failure occurred.

"excellent" – 200 or more

"good" – 150 to 200

"acceptable" – 100 to 150

25 "bad" – less than 100

Resistance to weather: A line of the conductive film of 0.8mm in width was baked on the glass substrate to leave to stand for 30 days in the atmosphere, and then the change of resistance value thereof was measured.

"excellent" – less than 5%

30 "good" – less than 10%

"acceptable" – less than 20%

"bad" – 20% or more

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Table 3

Sample Number	Resistance value ( $\Omega$ )	Solderability	Adhesibility (kg/4mm <sup>2</sup> )	Resistance to abrasion	Resistance to weather
1 *	21.7	acceptable	0.7	bad	excellent
2 *	7.8	excellent	0.5	bad(delaminated)	acceptable
3	26.0	good	3.2	acceptable	excellent
4	10.3	excellent	4.5	excellent	excellent
5 *	32.9	bad	-	bad(delaminated)	excellent
6	5.5	excellent	5.2	excellent	excellent
7	3.7	excellent	3.9	good	excellent
8 *	3.3	bad	-	bad	bad
9	2.1	excellent	3.0	acceptable	good
10 *	2.0	excellent	3.9	bad	bad
11	4.2	acceptable	4.4	good	excellent
12	1.5	excellent	2.8	acceptable	acceptable
13 *	Paste is not obtained; Application is not possible				

The samples with the mark \* in Tables 2 and 4 are out of the scope of the present invention. The rest falls in the scope of the present invention.

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As set forth above in Examples, the present invention obviously possesses the excellent characteristics which provides sufficient resistance to abrasion, resistance to weather, and solderability, and by which it is possible to obtain the value of the adhesibility to the terminal sufficient for practical use.

10

Patent Applicant:

Murata Manufacturing Co., Ltd.